

Theory of pump-probe experiments of metallic metamaterials coupled to the gain medium

Zhixiang Huang,^{1,2} Th. Koschny,¹ and C. M. Soukoulis^{1,3}

¹*Ames Laboratory and Dept. of Phys. and Astronomy, Iowa State University, Ames, Iowa 50011, U.S.A.*

²*Key Laboratory of Intelligent Computing and Signal Processing, Anhui University, Hefei 230039, China*

³*Institute of Electronic Structure & Laser, FORTH, 71110 Heraklion, Crete, Greece*

(Dated: March 1, 2013)

We establish a new approach for pump-probe simulations of metallic metamaterials coupled to the gain materials. It is of vital importance to understand the mechanism of the coupling of metamaterials with the gain medium. Using a four-level gain system, we have studied light amplification of arrays of metallic split-ring resonators (SRRs) with a gain layer underneath. We find that the differential transmittance $\Delta T/T$ can be negative for SRRs on the top of the gain substrate, which is not expected, and $\Delta T/T$ is positive for the gain substrate alone. These simulations agree with pump-probe experiments and can help to design new experiments to compensate the losses of metamaterials.

PACS numbers: 42.25.-p, 78.20.Ci, 41.20.Jb

The field of metamaterials has been spectacular experiments progress in recent years [1–3]. The mostly metamaterial are metal-based nano-structure and eventually suffering from the conductor losses at optical frequencies, which are stills orders of magnitude too large for the realistic applications. In addition, metamaterial losses become an increasingly important issue when moving from multiple metal-based metamaterial layers to the bulk case [3]. Thus, the need for reducing or even compensating of the losses is a key challenge for metamaterial technologies. One promising way of overcoming the losses is based on introducing the gain material to the metamaterial. The idea of combination of a metamaterial with an optical gain material has been investigated by several theoretical [4–7] and experimental studies [8–12]. From the experiments point of view, the realistic gain can be experimentally realized with fluorescent dyes [8], quantum dots [9, 10] or semiconductor quantum wells [11, 12]. All these loss-compensation are mainly attributed to the coupling between metamaterial and the gain medium. Without sufficient coupling, no loss-compensation can happen, nor can the transmitted signal be amplified. Therefore, it is of vital importance to understand the mechanism of the coupling between metamaterial and the gain medium. In addition, these ideas can be used in plasmonics to incorporate gain [13, 14] to obtain new nano-plasmonic lasers [15, 16].

In this Letter, we present a systematic theoretical model for pump-probe experiments of metallic metamaterials coupled with the gain material, described by a generic four-level atomic system. We describe the dynamical processes in metamaterials with gain, and increasing the gain changes the metamaterials properties and we need to have self-consistent calculations [4–6] to reach a steady state. The pump-probe results affecting the time dependence of the population inversion and the electric field enhancement that increases the effective gain. We observe differential transmittance signals from the coupled system that are larger than for the bare gain. Furthermore, we observe a more rapid temporal decay of the differential transmittance signal for the coupled system compared to the bare gain. Both effects indicate substantial local-field-

enhancement effects, which increase the effective metamaterial gain beyond the bare gain, leading to a significant reduction of the metamaterial's losses.

We model the dispersive Lorentz active medium using a generic four-level atomic system. The population density in each level is given by N_i ($i=0,1,2,3$). The time-dependent Maxwell's equations for isotropic media are given by $\nabla \times \mathbf{E}(r, t) = -\partial \mathbf{B}(r, t)/\partial t$ and $\nabla \times \mathbf{H}(r, t) = \partial \mathbf{D}(r, t)/\partial t$, where $\mathbf{B}(r, t) = \mu\mu_0 \mathbf{H}(r, t)$, $\mathbf{D}(r, t) = \epsilon\epsilon_0 \mathbf{E}(r, t) + \mathbf{P}(r, t)$ and $\mathbf{P}(r, t)$ is the dispersive electric polarization density that corresponds to the transitions between two atomic levels, N_1 and N_2 . The vectors \mathbf{P} introduces gain in Maxwell's equations and its time evolution can be shown to follow that of a homogeneously broadened Lorentzian oscillator driven by the coupling between the population inversion and external electric field [17]. Thus, \mathbf{P} obeys the equation of motion

$$\frac{\partial^2 \mathbf{P}(r, t)}{\partial t^2} + \Gamma_a \frac{\partial \mathbf{P}(r, t)}{\partial t} + \omega_a^2 \mathbf{P}(r, t) = \sigma_a \Delta N(r, t) \mathbf{E}(r, t)$$

where Γ_a stands for the linewidth of the atomic transitions at ω_a , and accounts for both the nonradiative energy decay rate, as well as dephasing processes that arise from incoherently driven polarizations. In the following simulations, this value is equal to $2\pi \times 20 \cdot 10^{12}$ rad/s. σ_a is the coupling strength of \mathbf{P} to the external electric field and its value is taken to be 10^{-4} C²/kg. The factor $\Delta N(r, t) = N_1(r, t) - N_2(r, t)$ is the population inversion between level 2 and level 1 that drives the polarization \mathbf{P} . In order to do pump-probe experiments numerically we first pump the gain material with a short, intense Gaussian pump pulse. After a suitable time delay we probe the structure with a weak probe pulse (see Fig. 1). In our model, an external mechanism pumps electrons from the ground state level N_0 to the third level N_3 using a gaussian pumping $P_g(t)$, which is proportional to the pumping intensity in the experiments. After a short lifetime τ_{32} electrons transfer non-radiative into metastable second level N_2 . The second level (N_2) and the first level (N_1) are called the upper and lower lasing levels. Electrons can be transferred from the upper to the lower lasing level by spontaneous and stimulated

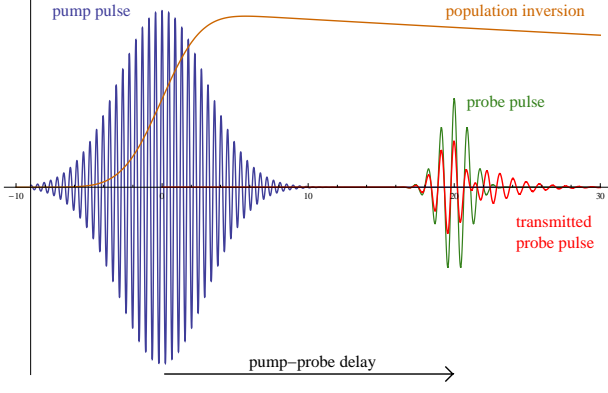


FIG. 1: (Color online) Schematic illustration of pump-probe experiments.

emission. At last, electrons transfer quickly and non-radiative from the first level (N_1) to the ground state level (N_0). The lifetimes and energies of the upper and lower lasing levels are τ_{21} , E_2 and τ_{10} , E_1 , respectively. The center frequency of the radiation is $\omega_a = (E_2 - E_1)/\hbar$ which is a controlled variable chosen according to the pump-probe experiments. The parameters τ_{32} , τ_{21} , and τ_{10} are chosen to be 0.05ps, 80ps, and 0.05ps, respectively. The initial electron density, $N_0(r, t = 0) = 5.0 \times 10^{23} \text{ m}^{-3}$, $N_i(r, t = 0) = 0 \text{ m}^{-3}$ ($i=1,2,3$). Thus, the atomic population densities obey the following rate equations:

$$\begin{aligned} \frac{\partial N_3(r, t)}{\partial t} &= P_g(t)N_0(r, t) - \frac{N_3(r, t)}{\tau_{32}} \\ \frac{\partial N_2(r, t)}{\partial t} &= \frac{N_3(r, t)}{\tau_{32}} + \frac{1}{\hbar\omega_a} \mathbf{E}(r, t) \cdot \frac{\partial \mathbf{P}(r, t)}{\partial t} - \frac{N_2(r, t)}{\tau_{12}} \\ \frac{\partial N_1(r, t)}{\partial t} &= \frac{N_2(r, t)}{\tau_{12}} - \frac{1}{\hbar\omega_a} \mathbf{E}(r, t) \cdot \frac{\partial \mathbf{P}(r, t)}{\partial t} - \frac{N_1(r, t)}{\tau_{10}} \\ \frac{\partial N_0(r, t)}{\partial t} &= \frac{N_1(r, t)}{\tau_{10}} - P_g(t)N_0(r, t) \end{aligned}$$

where Gaussian pump $P_g(t) = P_0 \times e^{-(\frac{t-t_p}{\tau_p})^2}$, with $P_0 = 3 \times 10^9 \text{ s}^{-1}$, $t_p = 6 \text{ ps}$ [18], and $\tau_p = 0.15 \text{ ps}$.

In order to solve the response of the active materials in the electromagnetic fields numerically, the FDTD technique is utilized [19], using an approach similar to the one outlined in [20].

The object of our studies is to present pump-probe simulations on arrays of silver SRRs coupled to single quantum wells [11, 12]. The structure considered is a U-shape SRRs fabricated on a gain-GaAs substrate with a square periodicity of $p = 250 \text{ nm}$ (see Fig. 2(a)). The SRRs is made of silver with its permittivity modeled by a Drude responds: $\epsilon(\omega) = 1 - \omega_p^2/(\omega^2 + i\omega\gamma)$, with $\omega_p = 1.37 \times 10^{16} \text{ rad/s}$ and $\gamma = 2.73 \times 10^{13} \text{ rad/s}$. The incident wave propagates perpendicular to the SRRs plane and has the electric field polarization parallel to the gap (see Fig. 2(a)). The corresponding geometrical parameters are $a = 150 \text{ nm}$, $h_d = 40 \text{ nm}$, $h_g = 20 \text{ nm}$, $h_s = 30 \text{ nm}$, $w = 50 \text{ nm}$, and $h = 75 \text{ nm}$. Fig. 2(b) shows

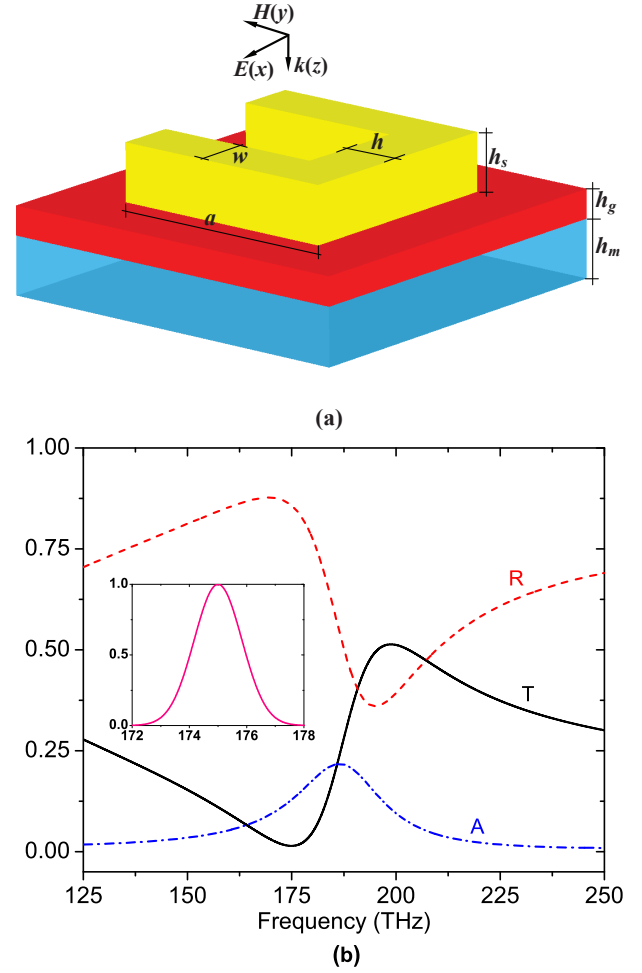


FIG. 2: (Color online) (a) Schematic of the unit cell for the silver-based SRRs structure (yellow) with the electric field polarization parallel to the gap. The dielectric constants ϵ for gain (red) and GaAs (light blue) are 9.0 and 11.0, respectively. (b) Calculated spectra for transmittance T (black), reflectance R (red), and absorptance A (blue) for the structure shown in Fig. 2(a). The insert shows the profile of the probe pulse with a center frequency of 175THz (FWHM = 2THz).

the calculated spectrum (without pump) of transmittance T , reflectance R , and absorptance A for the structure shown in Fig. 2(a). The resonant frequency is around 175THz, and we refer to the resonant frequency according to the dip of the transmittance. In our analysis, we first pump the active structure (see Fig. 2(a)) with a short intensive Gaussian pump pulse, $P_g(t)$, (see Fig. 3, top panel). After a suitable time delay (i.e. the pump-probe delay), we probe the structure with a weak Gaussian probe pulse with a center frequency close to the SRRs resonance frequency of 175THz. Typical examples for the spatial distribution of electric field an gain are shown in [21]. The incident electric field amplitude of the probe pulse is 10V/m, which is well inside the linear response regime. Then, we can Fourier transform the time-dependent transmitted electric field and divide by the Fourier transform of the in-

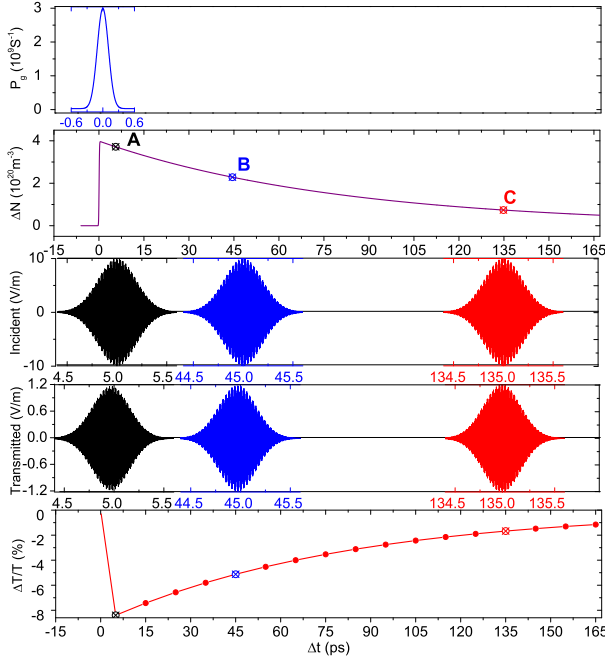


FIG. 3: (Color online) Schematic of the numerical pump-probe experiments for the case on resonance. From the top to the bottom, each row is corresponding to the pump pulse, population inversion, incident signal (with time delays 5ps, 45ps and 135ps), transmitted signal, and differential transmittance $\Delta T/T$. It should be mentioned here the incident frequency of the probe pulse is 175THz with FWHM of 2THz and is equal to the SRRs resonance frequency.

incident probe pulse to obtain the spectral transmittance of the system as seen by the probe pulse. Additionally, we obtain the total pulse transmittance by dividing the energy in the transmitted pulse by the energy in the incident pulse, integrated in time domain. We define the differential transmittance, $\Delta T/T$, by taking the difference of the measured total plus transmittance with pumping the active structure minus the same without pumping and dividing it by the total plus transmittance without pumping. This differential transmittance is a function of the pump-probe delay. The bottom panel in Fig. 3 gives a differential transmittance $\Delta T/T$ which is negative. This result was not expected, and we need to understand this behavior, which agrees with the experiments [11, 12].

Fig. 4 gives an overview over the results obtained for the case of the SRRs on resonance, i.e., $\omega_a = 2\pi \times 175 \times 10^{12}$ rad/s. Data for the structure in Fig. 2(a) (left column in Fig. 4) and for the bare gain case (right column in Fig. 4) without the SRRs on top is shown. For parallel polarization, the light does couple to the fundamental SRRs resonance, for perpendicular polarization it does not. The probe center frequency decreases from top (179THz) to bottom (169THz). Note that the width of the probe spectrum is 2THz (see the insert of Fig. 2(a)). Hence, the data have been taken with 2-THz spectral separation. Inspection of the left column shows a rather different behavior for the SRRs with gain compared to the bare gain case. While the bare gain always delivers positive $\Delta T/T$ sig-

nals below +0.16% (right column) over the whole probe spectrum. The sign and magnitude of the signals change for the case SRRs with gain. Under some conditions, $\Delta T/T$ reaches values as negative as -8.50% around $f_{probe} = 175$ THz. Additionally, we may also get positive $\Delta T/T$ at the very edges of the probe range (see left column in Fig. 4). If we turn to the case of perpendicular polarization case, no distinct change between the pump-probe results on the SRRs (not shown in Fig. 4) and the bare gain (right column in Fig. 4), neither in the magnitude nor in the dynamics of the $\Delta T/T$, can be detected.

We argue the distinct behavior can be attributed to the strong coupling between the resonances of the SRRs and the gain medium. The negative $\Delta T/T$ are not as we expected at first glance: the pump lifts electrons from ground state to an excited state so that the absorption of the probe pulse is reduced, leading to an increase of transmission. This is not the whole story. The reason lies in the fact that with pump we not only affect the absorption, but disturb the reflection of the structure, resulting in the mismatching of the impedance. Furthermore, we observed either increasing or decreasing tendency for the case of on resonance as shown in Fig. 4. All those behaviors can be explained by the competing of the weak gain resonance and the impedance mismatching between pump and without pump cases. We will explore the underlying mechanism below. Fig. 5 shows the

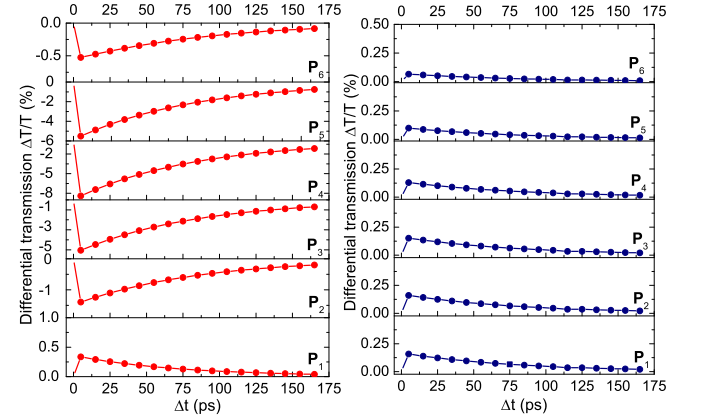


FIG. 4: (Color online) Time domain numerical pump-probe experiments results for the SRRs that is nearly on-resonant with the gain material. The left column corresponds to the parallel probe polarization with respect to the gap of the SRRs, the right column is the case for bare gain material, i.e., without SRRs on the top of the substrate. The width of the probe signal is 2THz with decreasing in the probe center frequency from 179THz for the top panel to 169THz for the bottom panel.

results for the difference in absorptance (ΔA), difference in reflectance (ΔR), their sum ($\Delta A + \Delta R$), and the difference in transmittance ($\Delta T = -(\Delta A + \Delta R)$) between pump ($P_0 = 3 \times 10^9 s^{-1}$) and no-pump using a wide probe (FWHM = 54THz) pulse with a fixed pump-probe delay of 5ps. As expected, we may observe a positive differential transmittance, $\Delta T/T > 0$, when we pump the gain, $\Delta A < 0$,

and if ΔR (impedance match) remains unchanged.

The results of Fig. 5 are obtained for pump-probe experiments with the probe frequency equal to the resonance frequency of the SRRs (175THz) at a pump-probe delay of 5ps; Results for longer pump-probe delays are shown in supplementary material [21]. Notice that ΔR is positive, ΔA is negative, and ΔT is also negative very close the the resonance frequency. If the probe center frequency moves away from

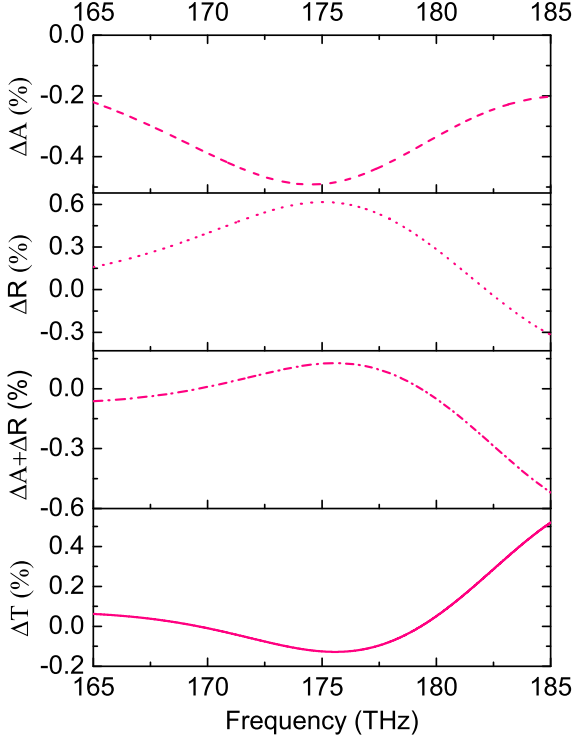


FIG. 5: (Color online) Frequency domain numerical pump-probe experiments results for the on-resonance case. Simulations results for the differences in transmittance (ΔT), reflectance (ΔR), and absorbance (ΔA) versus frequency.

the SRRs resonance frequency, the negative $\Delta T/T$ decreases in magnitude and finally $\Delta T/T$ becomes positive. These results are shown in Fig. 6 and agree with experiments [11, 12]. If we can increase the magnitude of the Gaussian pump pulse $P_g(t)$ to $5 \times 10^{10} s^{-1}$ and we repeat the pump-probe experiments, $\Delta T/T \approx -100\%$ at resonance frequency, 175THz. If we increase the pump amplitude further to $10^{11} s^{-1}$ we can compensate the losses. However, such pump intensities are unrealistic experimentally [21]. In conclusion, we have introduced a new approach for pump-probe simulations of metallic metamaterials coupled to gain materials. We study the coupling between the U-shaped SRRs and the gain material described by a 4-level gain model. Using pump-probe simulations we find a distinct behavior for the differential transmittance $\Delta T/T$ of the probe pulse with and without SRRs both in magnitude and sign (negative, unexpected, and/or positive). Our new approach has verified that the coupling between the

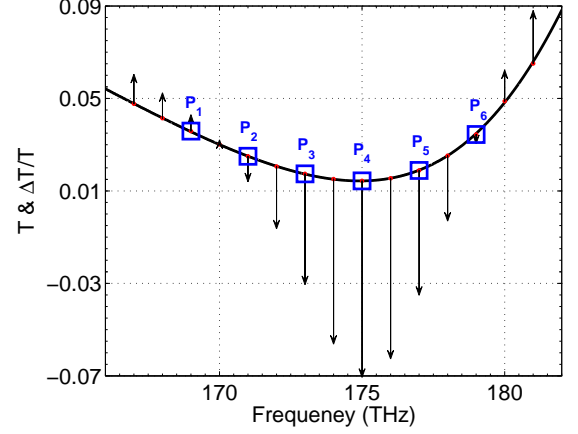


FIG. 6: (Color online) The transmittance T (without pump, solid line) and the on-resonance differential transmittance $\Delta T/T$ results (vector arrow). The direction and the length of the arrow stand for the sign and the amplitude of $\Delta T/T$, respectively. The squares from P_1 to P_6 correspond to the frequency of probe pulse ranging from 169THz to 179THz with uniform step of 2THz.

metamaterial resonance and the gain medium is dominated by near-field interactions. Our model can be used to design new pump-probe experiments to compensate the losses of metamaterials.

Acknowledgements

Work at Ames Laboratory was supported by the U. S. Department of Energy (Basic Energy Science, Division of Materials Sciences and Engineering) under contract No. DE-ACD2-07CH11358. This work was partially supported by the European Community FET project PHOME (No. 213390) and by Laboratory-Directed Research and Development Program at Sandia National Laboratories. The author Zhixiang Huang gratefully acknowledges the support of the National Natural Science Foundation of China (No. 60931002, 61101064), Distinguished Natural Science Foundation (No. 1108085J01) and Universities Natural Science Foundation of Anhui Province (No. KJ2011A002).

-
- [1] V. M. Shalaev, Nat. Photon. **1**, 41 (2007).
 - [2] C. M. Soukoulis, S. Linden, and M. Wegener, Science **315**, 47 (2007).
 - [3] C. M. Soukoulis and M. Wegener, Science **330**, 1633 (2010); Nature Photon. **5**, 523 (2011).
 - [4] A. Fang, Th. Koschny, and C. M. Soukoulis, Phys. Rev. B **82**, 121102(R) (2010).
 - [5] S. Wuestner *et al.*, Phys. Rev. Lett. **105**, 127401 (2010); S. Wuestner *et al.*, Phil. Trans. Royal Soc. A **369**, 3525 (2011).
 - [6] A. Fang, Z. Huang, Th. Koschny, C. M. Soukoulis, Opt. Express **19**, 12688 (2011).

- [7] Y. Sivan *et. al.*, Opt. Express **17**, 24060 (2009).
- [8] S. Xiao *et. al.*, Nature **466**, 735 (2010).
- [9] E. Plum *et. al.*, Optics Express **17**, 8548 (2009).
- [10] K. Tanaka *et. al.*, Phys. Rev. Lett. **105**, 227403 (2010).
- [11] N. Meinzer *et. al.*, Opt. Express **18**, 24140 (2010).
- [12] N. Meinzer *et. al.*, Appl. Phys. Lett. **99**, 111104 (2011).
- [13] D. J. Bergman and M. I. Stockman, Phys. Rev. Lett. **90**, 027402 (2003).
- [14] M. I. Stockman, Nat. Photon. **2**, 327 (2008).
- [15] R. F. Oulton *et. al.*, Nature **461**, 629 (2009).
- [16] M. A. Noginov *et. al.*, Nature **460**, 1110 (2009).
- [17] A. E. Siegman, Lasers (Hill Valley, California, 1986). See chapters 2, 3, 6, and 13.
- [18] The pumping rate is equivalent to a pump intensity. The pump power density is equal to $\hbar\omega_a P_g N_0$, and the pump intensity $I_p = (\text{pump power})/(\text{surface area}) = \hbar\omega_a P_g N_0 (\text{volume})/(\text{surface area}) = \hbar\omega_a P_g N_0 d$ and d is the thickness of the gain layer. If we use the numbers of our simulations, $P_g = 3 \times 10^9 \text{ s}^{-1}$, $N_0 = 5 \times 10^{23} \text{ m}^{-3}$, $\omega_a = 2\pi \times 175 \text{ THz}$, and $d = 20 \text{ nm}$, then $I_p = 3.5 \text{ W/mm}^2$.
- [19] A. Taflov, Computational Electrodynamics: The Finite Difference Time Domain Method (Artech House, London, 1995). See chapters 3, 6, and 7.
- [20] A. Fang, Th. Koschny, and C. M. Soukoulis, J. Opt. **12** 024013 (2010).
- [21] See Supplemental Material at [URL will be inserted by publisher] for additional details of the simulations and a discussion of overcompensation of the loss.